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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Development and impact of hooks of large droplet concentration on remote southeast Pacific stratocumulus" *by* R. C. George et al.

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Responses to Review 1

Reviewer: General Comments: This is an interesting and useful model study of the impact of pollution aerosol transported over the SEP during the VOCALS experiment and the characterization of model aerosol entrainment in modifying SEP cloud features. It is well suited to ACP and will be of interest to the atmospheric science and radiation community. This paper shows that WRFchem can capture the nature and transport of some of the aerosol layers observed above cloud during VOCALS including the inïňĆuence of the Santiago pollution on cloud structure (hooks) and CCN. Important re-

sulting inïňĆuences of this FT pollution are estimated for droplet number, cloud albedo and cloud fraction. The paper also points out discrepancies found in the model when used for this objective for select cases. However, direct observations during VOCALS revealed other examples of FT pollution transport and cloud interactions than those examined in these select cases [eg. Combustion Aerosol, Entrainment and Clouds in the VOCALS RegionCLIVAR Exchanges, v15, 2, 2010, Clarke et al.]. Moreover, the model did not yield consistent results for all of the 3 selected "hook" cases, so the generality of the conclusions regarding these aerosol/cloud interactions is not clear. The paper could be improved through a more detailed discussion/assessment of the model uncertainties and more complete investigation of the nature of identiïňĄed disagreements. This could also be furthered by taking greater advantage of the in-situ aircraft data available.

Although most of the approach and the modeling is communicated well, there are a number of places the presentation can be improved. Some are simple changes in sentence structure but others are more substantive. Generally, more quantitative statements could be employed throughout and greater discussion of uncertainties and their impacts upon conclusions. SpeciïňĄc issues/comments are outlined below. Although I like the thrust of the paper and I support publication, I feel that most of these comments need to be addressed before the paper is accepted for ACP.

Authors: The authors would like to thank Tony Clarke for his thorough review of this work and insights into its implications and suggestions for improvement. We appreciate his support for this publication and hope that he finds our responses to his concerns reasonable. He brings up several good points, some of which we address with changes to the paper, and some we acknowledge are outside the scope of this work that would be good problems for the future. It was not our intention to exclude his contribution towards understanding that the free troposphere can be an important source of stratocumulus cloud property variability. We have now added a reference to his CLIVAR Exchanges article, cited some of his previous work on the importance of the free troposphere as an aerosol source for the remote marine boundary layer, and added him to the acknowledgements because his ideas about these processes informed our understanding of the SEP. While our study primarily focuses on particular features over the remote ocean, we look forward to seeing his more comprehensive work on entrainment of pollution from the FT over the southeastern Pacific.

The following text has been added to the paper's introduction to acknowledge the contribution of Tony's work to inform this study: " …direct advection in the MBL from coastal sources; episodic transport in the FT and entrainment into the MBL (Huneeus et al., 2006; Clarke et al., 2010a). FT sources of CCN are important in many locations (Clarke and Kapustin, 2010). The Lagrangian evolution of MBL aerosol properties in stratus regions is strongly influenced by the composition of the overlying FT air; when clean serves to dilute the MBL (Clarke et al., 1996), and nucleation from convective cloud outflow and long range transport can provide sources of new particles introduced to the MBL via entrainment (Clark et al., 1998; Wood et al., 2000; Clarke et al., 2013). Aerosol observations and analysis (Clarke et al. 2010ab), revealed layers of pollution aerosol above cloud transport from South America in some cases. Active entrainment of pollution aerosol was observed at sizes effective as CCN in VOCALS stratus clouds (Clarke et al, 2010a,b) similar to previously documented influences from long range transport of pollution and entrainment found over the North Pacific (Clarke et al., 2001). "

One point we feel it important to clarify before providing detailed responses to individual comments is that this study is not intended to be a model evaluation study. We use limited observations to note what general aspects of the model do and do not match well and to point out how difficult hook features in cloud droplet concentration are to simulate, but it was not our intention for the main focus to be model performance. Two other papers rather rigorously evaluate WRF-Chem simulations in the SEP against observations (Yang et al., 2011, Saide et al, 2012). Although the model does not perfectly match observed hook scenarios, it is able to reproduce some of the observed features.

We then use the model to assess the likely impacts that this aerosol transport to the remote ocean might have on the clouds and radiation, and explore the meteorological mechanisms that support hooks. Although the model does not reproduce the latter two cases as well as the first, these are substantially weaker events as discussed. A detailed evaluation of the model's ability to capture the correct aerosol size distributions, cloud top entrainment, etc. noted in a number of the reviewer's suggestions would be a useful piece of analysis that could be carried out in a separate paper. However, we believe that trying to do so within this paper would obscure our main message, namely to describe the salient features involved in producing episodes of pollution transport that reach the remote ocean. We try to cover first order effects of aerosols, chemistry, meteorological forcing and transport and therefore no individual topic can be explored in great detail without the paper becoming unacceptably long. The lack of a strong focus on comparison with in situ data reflects the noted lack of sampling of large CCN concentrations in the FT that entrain into the MBL in the remote ocean. We believe that this primarily reflects misfortune in timing in the VOCALS flights, which were not designed to sample hook-like features.

A second point of necessary clarification is that our primary focus is on the causes of specific events of high cloud droplet concentration in the remote ocean and is not a complete study of entrainment of free tropospheric aerosols into the SEP stratocumulus. The work was motivated by the temporal variability in droplet concentration and where its contribution to albedo variability is greatest. Thus the focus of this paper was not driven by the question of how free tropospheric aerosols influence cloud properties, but rather what mechanism is driving the observed cloud microphysical changes.
Specific comments:

L13. Reviewer: "..the model suggests that high concentrations of …" Because pollution aerosol with high number but low mass (compared to the MBL) were regularly observed above cloud during VOCALS it is not informative to say the WRFchem model "suggests" their presence. It would be more relevant to say something like "…the model captures the high concentrations of pollution aerosol observed above cloud during VOCALS. . ."

Authors: Thank you, agreed, though this sentence is referring specifically to the FT CCN source aerosols for the hook events in the remote ocean, which was not seen in aircraft legs means over the remote ocean (besides 1 leg described for Hook 3). We add "in agreement with near-coast VOCALS measurements of polluted layers in the FT" to address this.

L16: Reviewer: ". . .originate mainly from a pulse of offshore ïňĆow that transports Santiago. . ." This may occur frequently and be true for the limited cases discussed in this paper but pollution was often observed above cloud for more than just the cases discussed in this paper and sometimes originated from very different sources. Rephrase

Authors: We are referring to the "FT source supplying the hook," which we've defined as a remote ocean feature. While there are other sources of CCN in the FT during REx, the FT source for hook cases appear to be most commonly related to Santiago emissions as variability in the synoptic meteorology can periodically favor the transport mechanism from this region. Cases of pollution measured in the FT during REx were generally not in the remote ocean and not associated with a hook feature downwind. But, indeed, it is possible to have transport from other regions as well, as demonstrated by Hook3 trajectories, which indicate transport from both the Santiago region and possibly a northern volcano. Nonetheless, this portion of the abstract is referring to Hook1. We agree this was not clear and have reworded to be specific to Hook1: "The aerosol particles in this hook originate mainly from a pulse of oïňĂshore ïňĆow that transports Santiago region (33–35âĄřS) emissions to the remote marine FT."

P2498 L19 Reviewer: What justiïňĄcation exists for this choice of the %S of 0.5? This is about twice the %S for VOCALS stratus as evident in the observed dry-size aerosol Hopple minimum. This choice will lead to larger absolute model numbers and a dif-

ferent sensitivity to diurnal ïňĆuctuations in %S (discussed later) than would normally occur. Given this choice has a direct impact in assessing CCN, I feel that authors need to clearly show model size distributions explicitly and indicate where the activation threshold for 0.5 %S (and 0.25 %S) would be expected. Were any runs done at %S less than 0.5 and, if so, how different were they? Given that this choice is fundamental to the results in the paper, this needs to be discussed accordingly.

Authors: CCN at 0.5 % was shown originally because as seen in Figure 8a, it closely corresponds to the droplet concentration during hook development. An important point here is that the model does not run the simulation at 0.5% SS, but rather internally computes a supersaturation and activation of particles from TKE and the modeled aerosol size distributions following Abdul-Razzak and Ghan (2000). The model also outputs diagnostic CCN variables at various prescribed supersaturations, and 0.5% is one of them. The CCN shown is a diagnostic output that does not impact the sensitivity of Nd to supersaturation. Just after the paper was submitted we found the WRF-Chem diagnostic output of CCN appeared to make erroneous mode size assumptions, so we have recalculated CCN for the new submission. The recalculation indicates that the CCN at 0.3% SS corresponds mostly closely to Nd. That said, the focus of this paper is not about precise details of the CCN and size distribution. Showing model size distributions would not change how we interpret the mechanism of hook development.

P2499 L4 Reviewer: "..midday retrievals reasonably match. . ." If this result does not merit a ïňĄgure then a more quantitative assessment of "reasonably" needs to be speciïňĄed in the text. How well do they match and in what way?

Authors:Thank you. This assessment comes from the cited Painemal et al. (2012) paper, and we now add a more specific assessment. Added: "The GOES and MODIS derived Nd from optical depth and effective radius retrievals have a strong correlation coefficient (0.88) at coincident times and a mean bias of 33.4 cm-3. This is sufficient for distinguishing events of high Nd in the remote ocean."

L21. Reviewer: "although accumulated trajectory errors could potentially account... This statement is premature and not an observation. A discussion of trajectory performance and associated errors is needed before it can be interpreted.

Authors: The trajectories are an estimate of the flow based on 3 hourly output from the model. We note that along the trajectory paths Nd increases over time, so we need to acknowledge that it is possible the errors in trajectory calculations are large enough to shift the estimated trajectories with respect to the developing cloud feature. That it is, back trajectories intending to follow the region of large Nd could accumulate large enough errors to shift outside the main the hook. It is not intended to be an observation, but is rather a reference to a potential reason for trajectories no longer tracking a high Nd MBL feature. The appendix explains how trajectories are computed and describes why and how there can be errors, and the appendix is referred to in the previous paragraph. We have added another reference to this appendix here to allow the reader to examine the details of this statement. A manuscript from our group that is currently in preparation explores in greater depth the potential trajectory location errors in the region.

P2500 L5 Reviewer: 5 It is statedâĂŤ "The two most western trajectories do not quite intersect the coast, the clearly connected nature of the observed hook suggest these locations have similar sources." However, if one looks at tracers like CO, black carbon and organics etc, these indicators are lower for these two outermost longitudes than the others, suggesting there is a real difference.

Authors: We do not know that tracers for CO, organics etc. would be lower in the further most points in this particular case. We do not have observations of this case. If they are indeed lower because they transported a larger distance/time, this doesn't negate the possibility of a similar source, just differences in the distance and time the source has been transported. In this statement we are trying to convey it is unlikely that the western end of the hook is on the one hand derived from natural marine sources when the rest of the hook is driven by continental sources.

L18 Reviewer: The authors say "...ïňĄre related aerosol are unlikely to cause the heavily polluted Nd...". However, the above mentioned trajectories further east did have these high Nd concentrations above cloud and had enhanced organic and black carbon concentrations. Most pollution has a combustion source (ïňĄre) as evident in the general relation observed aerosol BC and CO during VOCALS.

Authors: Good point. We meant to say 'biomass burning' rather than 'fire related.' This has now been corrected.

L24 Reviewer: I do not think the data mentioned demonstrates a "...lack of an FT source" or perhaps better stated "lack of an FT supply of pollution derived CCN". This kind of FT/MBL comparison of CCN and potential FT contributions requires great care and cannot be explored without a discussion of uncertainties, winds and wind shear, subsidence and timing. Model layers and model invsrsion heights have to match reality too. Observed above-cloud layers were often patchy, variable and often thin. Infrequent CCN measurements in the FT also limit the assessment of these aerosol ïňĄelds in the FT. Also, for an entrainment rate of say 0.4 cm/s would take 10 hours to entrain a 100m layer. Hence, what was above the inversion during the past 10's of hours is more important than what was observed above cloud but yet to be entrained. Given the wind directional shear often present across the inversion, it is the upwind time-integrated inïňĆuence of the FT layers on the MBL that controls the resulting MBL concentration. This "exposure" is something the model can be interrogated to provide. On this ïňĆight, enhanced pollution aerosol at CCN sizes in the 200-300 /cm3 were measured above cloud along 20S between 80W and 77W (CCN measurements were limited). In general, more effort to compare with other in-situ data should be made and some accounting for possible uncertainties should be discussed before assuming the above cloud data at west end of ïňĆight path implies the MBL there had not been inïňĆuenced by prior entrainment. Also, I would avoid using "an FT source" as it is not a source of the aerosol in the same sense as say Santiago. Maybe an FT aerosol layer of enhanced CCN - or something like that.

Authors: We agree that the relationship of FT CCN to MBL Nd involves a number of complex processes and there is a timing component. We do discuss many of the complexities later in the paper. Here, we just noted we did not happen to measure elevated FT CCN on the available flight relevant to Hook1. This of course does not mean there is not one upstream. Indeed, our main conclusions, if true, must invoke one. We did not intend to imply that the observations demonstrate the lack of an entrainment source, but rather that they are unable to provide positive support that there is for this case. This further motivates using a model to look at the issue.

We have rephrased the text to reduce the emphasis on needing to measure a coincident FT source as follows: "Although any CCN in the FT that influences Nd must have been entrained upstream, there are no observations of a continuing polluted FT CCN at flight locations. It is thus difficult with REx observations alone to establish the source of hook CCN."

We are a little confused about your result that on this flight the aircraft measured 200-300 cm-3 CCN in the FT? We checked the XCNHot measurements from the C-130 and offshore leg mean values were within 100-200 cm-3. Individual measurements of higher CCN concentrations we do not feel are indicative of the large scale source that provides particles for hook Nd. Thin layers of large CCN tend to be diluted when entrained into the boundary layer, so only sufficiently large (thickness and horizontal extent) aerosol layers might enhance the Nd downstream. For example, to double the CCN concentration in the boundary layer from 100 /cm3 to 200 /cm3 given 300 /cm3 in the FT would require an FT layer that is the same thickness as the PBL, i.e. 1.5 km. Thin layers would not be sufficient.

Again, we do not meant to imply that flight measurements indicate the MBL has not been influenced by entrainment. It is not obvious what other measurements exist that would add more to the story before looking at model results. This section was intended to motivate using the model to enhance the story told by observations.

P2501 L10 Reviewer: What is model resolution in the vertical just above the inversion where transport and entrainment needs to be resolved in this paper and what does this imply for uncertainties? For example a 50m layer resolution could be associated with an uncertainty of about 5 hours for an entrainment rate of say 0.4 cm/s.

Authors: The vertical resolution near the inversion level is $\sim$200m and may be a large source of error. This does not necessarily mean the uncertainty associated with entrainment time is 20 hours because the PBL scheme is designed to handle coarse vertical resolution stratocumulus topped boundary layers. It is not clear that the provided example estimate of the error in entrainment fully characterizes the sources of model error in producing the hook. The entrainment rate is one such possible error, but likely more important are errors in the vertical extent of the pollution that advects offshore, for which we have no observational constraints. Large subgrid variability in topography also impacts the timing and elevation of offshore transport of pollution. The model is complex, with many interacting components, and a full characterization of the model uncertainties is beyond the scope of what we are trying to show in this paper. Yang et al. 2011 (Atmos. Chem. Phys.,11, 11951-11975, 2011) addresses a number of these issues. The model currently does not provide entrainment rate information, and given we only have saved 3 hourly model output it is not possible to estimate the entrainment rate accurately for our simulations with model output fields. To quantify the entrainment error we would need to run the model again and add an entrainment rate diagnostic output making use of the 3 minute time resolution information (similar to what was done in Yang,Atmos. Chem. Phys.,11, 11951-11975, 2011). Without doing this, we cannot tell if the entrainment error estimate really should be based on 0.4 cm/s. It is possible that the model entrainment rate incorporates vertical resolution information. Certainly the vertical model resolution may be one reason for some of the mismatch between observations and model, but given the PBL parameterization is designed to simulate stratocumulus layers well in coarse resolution models, it is not obvious that this is a major problem. Quantifying the uncertainties in entrainment rate would be more useful if we were doing more of a model evaluation study and trying to quantify more rigorously the model capabilities.

L19 Reviewer: factor of 100? Warrants a little more information. Seems drastic, is there some problem here, maybe model removal is way off?

Authors: The reduction of Lima emissions was drastic, and is an area of improvement for future simulations, but as noted, it does not impact the region we are studying. Model removal may be poorly represented (actually overestimated, Saide et al., 2012), but it is also possible transport or entrainment is incorrect in the northern part of the domain, or a number of other model errors could be causing this. It is also possible the Lima emissions are too large – when we compared to other emission estimates the VOCA emissions appeared to be an order of magnitude larger. In the model simulation with original VOCA Lima emissions the column SO2 does not compare well with OMI satellite measurements (Carn et al, 2007) and leads to very large droplet concentrations downwind of Lima. Saide et al. (2012) notes an overestimate of removal, which is the opposite sign of the bias leading to large Nd. So, future work would need to improve the Lima emissions estimate and address whether model physics are incorrectly processing these emissions.

P2502 L18 Reviewer: 8 How consistent are these model assumptions with the DMS ïňĆux assessment made by Yang et al. (Atmos. Chem. Phys., 11, 5079–5097, 2011) for the VOCALS region at this time?

Authors: The other Yang 2011 paper (Atmos. Chem. Phys.,11, 11951-11975, 2011) has an in-depth analysis of the assumed DMS flux suggested for use in the VOCAL model intercomparison experiments and compare DMS concentrations to Ron Brown measurements. They find that DMS concentrations are too large likely due to an overestimate of the transfer velocity, but a low bias in sulfate indicates either a problem in DMS oxidation or too strong of sulfate removal. We use the same DMS emissions and our results similarly overestimate DMS and underestimate sulfate.

L23 Reviewer: As mentioned earlier, size distributions are critical to modeling CCN and

they can be compared directly with measurements. The WRFchem model distributions mentioned need to be shown in a ïňĄgure along with the sizes expected to be activated at the assumed 0.5%S. This is particularly true as later diurnal ïňĆuctuation in %S and activation are discussed in some detail. This ïňĄgure can be then referenced directly in the context of sensitivity of the model to diurnal ïňĆuctuations in %S.

Authors: The model does not assume supersaturation is 0.5%, that is just the CCN we have shown (see response to P2498 L19). If we had assumed a 0.5% SS we would agree evaluation of size distributions would be a useful addition, but given that this field does not influence the relationship between Nd and supersaturation, we believe it would not add new information to our interpretation. We do show sizes of the accumulation mode aerosols, which are the most relevant for Nd.

P2503 L16 Reviewer: "...magnitude underestimated by 10-30%." This seems odd as the %S used in the model is higher than evident from the MBL size distributions (Hoppel minimum). Later, when comparisons to other measurements and albedo etc. is made, reference to this uncertainty should be included in interpretation

Author: The model supersaturation is not 0.5% (see response to P2498 L19). The actual model supersaturation is dependent on the model state within the activation parameterization, which is not output before being altered by other physics parameterizations and thus to evaluate how well model supersaturations compare to observed supersaturations we would need to alter the model to output this. Given the model takes quite a bit of time to compute, we do not take that step here.

L18 Reviewer: Comparisons to Saide et al. are mentioned but not for DMS. This is argued to be overestimated by Saide et al. in WRFchem. More generally, this section merits a more complete discussion of similarities and differences in models and performance (and with observations) rather than alluding to "reasonably reproduces REX mean conditions". It is not clear what that means. Is this "reasonable" in the MBL or in the FT or both? How reasonable is it? More importantly, some discussion of how the differences (or uncertainties) in absolute or relative quantities might impact (or not) the assessment of model CCN in the FT or MBL should be included and/or to what extent these differ from the Saide et al. WRF performance assessment for various key model parameters. How well (quantitiatively) does this WRFchem get the inversion height and cloud top correct as this will affect reliability of timing inïňĆuences for advected FT aerosol interactions with clouds compared to reality. What are implications of any key differences found?

Authors: We do not compare all possible fields with observations or Saide et al and Yang et al because a) it would make the paper too long and unfocused, b) this is not intended to be a model intercomparison study. Such a study would merit its own paper and c) Saide and Yang already presented very thorough analysis of WRF-Chem capabilities in this region, and our model compares to observations very similarly to these other studies. Additionally, a comparison between several models and observations will be featured in the upcoming VOCA model intercomparison paper, so we do not intend to pre-empt that. We realize we did not emphasize this point, and the text has been modified to make this more clear: "Although our simulations employ slightly different parameterization options and resolution, it compares similarly with REx mean conditions as these other studies so we do not reproduce their analysis here."

We've now also added text in section 3.2 to illuminate some of the suggested comparisons, but again implications of model errors is not the focus of this study. We are not generally concerned with reproducing the observed absolute values of CCN within the model simulation perfectly, but rather use CCN to contrast polluted versus clean times/places.

P2504 L7 -10 Reviewer: This shift of several degrees in model/observed behavior merits a greater discussion in the context of uncertainties. Horizontal and vertical uncertainties are coupled in a 3-D subsiding aerosol ïňĄeld and need more discussion. For example, what does horizontal displacement error imply in terms of altitude uncertainties for when or where a pollution layer will encounter the inversion and start to

have an effect on CCN? This discussion is important for this paper and the strategy for choosing when or whether or over what scales the in-situ measurements or satellite retrievals can be compared to a model output etc

Authors: Please see the second paragraph in this review response. There are many convolved uncertainties in this complex model, and a full characterization of these impacts is beyond the scope of this work. A separate paper could be written to establish relevant scales for comparison of model and observations based on the model resolution. It is a fascinating, important topic, but beyond the intentions of this paper.

P2506 L20-25 Reviewer: Here again reference needs to be made to assumed size distributions and the CCN activated under the inïňĆuence of different %S. Are these model distribution behaviors consistent observations? Similarly, is sensitivity to TKE in the model consistent with that expected for observed sizes and variability actual typical cloud supersaturation (eg. width of Hoppel minimum)

Authors: See responses to P2498 L19. The model does not have assumed size distributions; there are 3 modes whose mean can change with fixed mode standard deviations. This is an interesting topic, but we only meant to point to the impact of changing supersaturation (by noting TKE changes) on Nd variability, not to characterize the uncertainty or discuss the size distribution comparisons to observations. To address this we would want to modify the model to output the supersaturations that activate droplets at minimum, which would take a large amount of time, when this particular aspect of the hook behavior is not central to the paper. The variability in Nd due to changing supersaturation is interesting, but as noted in the paper the addition of continental aerosols to create the hook is the more important effect we focus on.

P2508 L1 Reviewer: "...only a small increase in MBL aerosol mass occurs as the FT aerosols are entrained...". IMPORTANT –If this is really what the model shows, I worry about how the model is handling mass conservation. Entrainment of air with lower aerosol mass concentrations can only dilute (lower) the MBL mass concentration and not increase it. Please explain

Authors: Good point. This was poorly explained and has now been fixed. The small increase in MBL mass is likely due to condensation of DMS derived SO2 on entraining and existing PBL aerosols.

L2 Reviewer: "Thus the FT is comprised of numerous small aerosol. . .". This is not a ïňĄnding as it was observed directly during VOCALS in the airborne aerosol size measurements. A more relevant concern is how well model and measured number distributions compare with model estimates for different air mass/aerosol types in the FT? Some evidence that model is getting aerosol sizes correct is needed.

Authors: This is a finding of the aerosols that contribute to hook cases in the remote ocean, we do not mean to imply that we had discovered that polluted aerosols are numerous and small. We cannot compare number and model number distributions for aerosol mass and types in the FT for this hook case because the aircraft did not measure the source the model is describing. A REx VOCALS domain comparison of aerosol number and types was carried out in the other WRF-Chem papers mentioned and we do not intend to reproduce their analysis in this paper.

P2509 L9-12 Reviewer:Please clarify discussion. In a typically divergent region with constant or increasing mean wind (typical) and with cloud base constant but with cloud top growing, it seems like signiïňĄcant entrainment of FT air is required for mass conservation. Or am I misreading the argument.

Authors: Good question. In this case we are arguing that weakening subsidence allows the cloud top to rise (for a fixed entrainment rate), but increasing stability likely reduces the entrainment and prevents the cloud base from rising as well. Of course, the reduced entrainment could offset the effect of the reduced subsidence but it does not have to. So a rise in cloud top and a flat cloud base is a reasonable outcome. The mass that allows the MBL top to rise must still come from entrainment, but it might be weaker than the REx-mean value.

P2512 L8-15 Reviewer: Here is another example of model uncertainty that will affect comparisons with measurements or satellite retrievals near locations of expected entrainment. It is good that such examples are included but it would be more useful to have uncertainties collected and discussed clearly in one section (such as pg. 2503) that then propagatesthem into uncertainties in some of the ïňĄndings reported.

Authors: This is another good point, but characterization of uncertainties requires very complex metrics and model-observation comparisons that are beyond what we are trying to show here. The transport over topography likely has errors, but observed wind datasets, such as reanalysis also contain topography induced errors. Characterizing the errors in entrainment rate due to inversion level vertical resolution is only one of many sources of uncertainty, and it is unclear whether the particular one suggested would be representative of all uncertainty or only comprise a small fraction of it. The model is not a perfect representation of reality, nor is it necessarily intended to be. It captures a feature of interest and error bars on its estimate are both difficult to estimate and will likely not change the interpretation of the important mechanisms affecting hook formation.

P2514 L19 Reviewer: Is this claimed rapid coagulation of Aitken mode number over 12 hr. and subsequent stabilization consistent with expected coagulation in the FT for these sizes and cncentratons? This appears to be too fast for a non-cloud environment. How did model size distributions change?

Authors: Initially we also thought that the Aitken mode reduction was too large to be explained by coagulation, but using the number concentration, size and time using Brownian coagulation kernel estimates from the Seinfeld and Pandis and Jacobson books, we computed estimates of number loss that were similar to that produced in the model. There is not a huge impact on the size distribution because Aitken mode particles commonly coagulate with accumulation mode particles (in this case the concentration is very high), reducing the number of Aitken aerosols, but changing the mean size of each mode very little. We added some text to explain this a little more clearly.

P2516 L1-10 Reviewer: There are certainly other, perhaps less pronounced, hooklike features present during VOCALS that also have pollution above clouds than the ones selected in this paper. Line 26 it indicates the model is not correctly simulating offshore advection. This implies that many examples or features may not be seen in the model or captured correctly for different sources.

Authors: The motivation for (as established in the Introduction of the paper) and intention of this study is to study cases of high Nd in the remote ocean. Indeed, there were many hook-shaped features during REx closer to the coast that may be related to FT transport, and we thought that these would fall into the territory of the paper you are writing on entrainment of FT aerosols.

L17 Reviewer: "The transition…". Is this transition in the MBL of FT? Authors: The hook is defined as an Nd feature, so we mean the MBL. We have added clarifying text.

L21 Reviewer: "...lack of FT support…" Again, what is above MBL for any measurement proïňĄles does not mean that this is what has been above it and entraining into it for the past day.

Authors: Thank you, the text incorrectly implied observations show there is no entraining FT pollution, we have rephrased.

P2517 L13-19 Reviewer: The authors should provide more complete discussion/clariïňĄcation/evaluation of the merits of their speculations on why WRFchem does not consistently produce hooks. Text Reason 1) As removal of CCN takes place in the MBL, if entrainment is the source then these should show up in the cloudiness changes before they can be removed by precip. Or are authors suggesting entrainment is suppressed? Text Reason 2) nucleation requires time and a precursor source strong enough to grow them to CCN sizes. Again, a comparison of model and measured size distributions and associated CCN in the plume seems warranted before speculation. Text Reason 3) Errors in altitude, latitude and magnitude of model ïňĆow were mentioned at several points in the text. As noted previously, a separate section that provides

a more quantitatively discussion of expected and observed uncertainties (eg. altitude) in the context of predicting CCN and hooks would be helpful. It is not constructive to speculate on these possibilities without following up. On Nov. 2, the in-situ measurements show pollution aerosol sizes at high concentrations in the MBL (not removed) out to beyond 80W. Given the extensive aerosol, cloud, chemistry,meteorological data etc. available on the C-130 one expects that model altitude ïňĄdelity and uncertainty could be better demonstrated/constrained and the likelihood of some of these reasons eliminated. Does the model get the inversion height correctly for these ïňĆights? etc..

Authors: We appreciate the great ideas, but would like to note the speculated reasons for model errors are not just guesses, but rather come from knowledge of what processes are represented in the model. The (1) reason why Hook2 is not reproduced well does not refer to entrainment, but rather the MBL component of transport. As noted earlier, MBL back trajectories follow large Nd back towards the coast, unlike Hook1. Thus this case may have a direct MBL transport component. Also, the model 3D trajectories indicate that entrainment of aerosols from the FT may be important before the remote ocean, that MBL transport extends high Nd to the remote ocean. Thus, if the model overpredicts MBL CCN losses, the source CCN for the hook may be processed before the hook reaches the remote ocean. (2) We do not have sufficient observations to compare observations and model size distributions in this case. The model hook does not reach the domain that was sampled by the aircraft. (3) When choosing a particular resolution to model a region, there are inherent uncertainties due to this choice, so this is not pure speculation, but rather a consequence of using a model. Actually quantifying these uncertainties is not a simple task, especially since there are not observations to constrain our expectation. On Nov 2 the aircraft measurements did go beyond 80ïĆřW, but our modeled hook does not reach the latitude of measurements, thus a detailed comparison with this data is not informative to hook representation.

Although we agree a model-observation evaluation study would be useful, again, that is not the intended purpose of this paper and an upcoming VOCA paper will address this. No regional or climate model has been shown to reproduce individual events, and it is not necessarily expected that any model will be able to do this well just because REx mean gradients are captured. The model is complex, and represents many processes that interact. A complete study of model discrepancies would require much more research and time, and is different focus from what we have shown in this study. Additionally the second two hooks explored were much weaker and less extensive features. If the model were to be run for longer and encounter more cases like Hook1 (which are seen in satellite observations), it would likely be able to reproduce these as well as Hook1.

Technical corrections

Authors:Thank you for the helpful technical questions, we appreciate having a fresh set of eyes take a detailed comb through the work.

Reviewer:Title could be made more clear Perhaps something like –Large droplet concentrations in arcs of cloudiness over the remote southeast PaciïňĄc: Their origin and impact.

Authors: Thank you for the great title suggestion. We have altered the title slightly to clarify.

P2494 L3 Reviewer: "hook shaped arcs" is redundant (–like "round circles"). Why not use just arcs throughout as they are less ambiguous in meaning?

Authors: The 'arc' or 'hook' debate was discussed much by the coauthors and we still think the term 'hook' provides the most clear description of the features we refer to. The term 'arc' could come up in other descriptions of geometric features, but 'hook' is less commonly used. We take your suggestion and replace 'arcs' with 'features' to remove redundancy in the shape description.

P2494 L18 Reviewer: change to "To provide pollution CCN that can sustain hooks..."

Authors: changed.

L23 Reviewer: change to " LWP also increases as the hook evolves over time...."

Authors: changed.

P2496 L22 Reviewer: change "topographic transport restrictions" to "transport restricted by topography"

Authors: changed.

P2543 Reviewer: Nice ïňĄgures (13,14) in principle but very hard to read numbers. Authors: Fig. 13 will be larger in the final edition (portrait in ACP vs landscape in ACPD). We have also added larger labels. If still hard to read, some zooming may be required.

P2545 Reviewer: Not clear this Fig is needed.

Authors: We focus in the text on the large scale meteorological conditions that influence aerosol transport, and this figure shows what is happening in the bigger picture to the subtropical high and the upper level flow as the hook aerosols are transported. The particular synoptic scenario is important and is what allows for offshore transport of aerosols to the remote ocean. The combination of the eastward shifting strengthening subtropical high behind an upper level trough is the reason Hook1 forms. Although we don't explore it in this paper, there is a strong association of shifts in the subtropical high with offshore flow (e.g. Rahn and Garreaud, 2010).

P2546 Reviewer: Switching to SO2 for 16c is confusing. At least show CCN (black) along with the SO2.

Authors: We would show CCN in black, but the concentrations of FT CCN are lower than the smallest threshold shown for CCN in the other panels and there would be no lines in our area of interest. At the time of c) in the model the concentrations of CCN in the FT are low, so only SO2 is high. This is one of the reasons described why the model may fail to reproduce this hook in the remote ocean. Since the polluted FT plume is mostly SO2 and has not nucleated to CCN, once entrained this SO2 tends to contribute to existing particles rather than add to the aerosol or droplet number.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 2493, 2013.